SCK•CEN
STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

RETROSPECTIVE DOSIMETRY FEASIBILITY STUDY FOR THE ATUCHA I NUCLEAR POWER PLANT

Background

The Reactor Pressure Vessel (RPV) of a nuclear power plant contains the nuclear fuel and is a primary loop barrier. The integrity of the RPV is a key component in the safety assessment of the plant. During reactor operation the RPV gradually degrades. The main damaging mechanism is the material embrittlement due to the neutron exposure. The embrittlement of the RPV steel is the limiting factor for the lifetime of the reactor. Therefore all modern nuclear power plants are equipped with a set of RPV surveillance capsules containing steel specimens of the same type of steel the RPV is made of (to investigate the material degradation) and neutron dosimeters (to determine the neutron exposure responsible for the degradation). The capsules are placed close to the RPV before the reactor start and dismantled and analysed on a regular basis during its lifetime. Surveillance capsules are ideally located near the beltline region (the section of the vessel wall closest to the fuel), at positions where the neutron flux is about two to three times higher than at the RPV wall. Such anticipated surveillance allows predicting the condition of the RPV.

Reactor dosimetry aims at determining the neutron fluences accumulated by the RPV wall during the reactor operational lifetime. This information can be deduced from the measured nuclide specific radioactivity of the irradiated neutron dosimeters (from the surveillance capsules) and the detailed irradiation history (including neutron spectrum calculations).

In some reactors no adequate RPV surveillance programme is available. In such cases one must rely on alternative techniques like e.g. ‘retrospective dosimetry’: steel specimens that are cut from structural material (like the RPV) are analysed and the neutron fluences that these specimens incurred are deduced. Even when routine dosimetry is available retrospective dosimetry can be a valuable technique to obtain complementary results. The RPV surveillance of the Argentinean reactor Atucha I is an example of a situation where retrospective dosimetry can be useful. Due to the lack of in-vessel positions, the surveillance capsules are positioned below the fuel elements resulting in irradiation conditions substantially different from those at the RPV beltline region. Since this does not allow a direct monitoring of the RPV neutron exposure, the dosimetry programme was complemented with an extensive 'ex-vessel' dosimetry campaign initiated in 1994. These results could be supplemented by retrospective dosimetry.

Retrospective dosimetry of fast neutron fluences is usually based on the reactions ^{54}Fe(n,p)^{54}Mn and ^{93}Nb(n,n')^{93m}Nb. For the first reaction a necessary requirement is that the irradiation and cooling times are not exceedingly long, the half life of ^{54}Mn being 312 days. The latter reaction can only be applied if a sufficient amount of niobium impurities is present in the steel.

In some cases retrospective dosimetry of thermal neutron fluences is also of interest. In Atucha I for example the surveillance capsules were positioned in a high thermal neutron fluence environment. Reactions that can be used for this purpose are for example ^{59}Co(n,γ)^{60}Co, ^{54}Fe(n,γ)^{55}Fe and ^{62}Ni(n,γ)^{63}Ni.

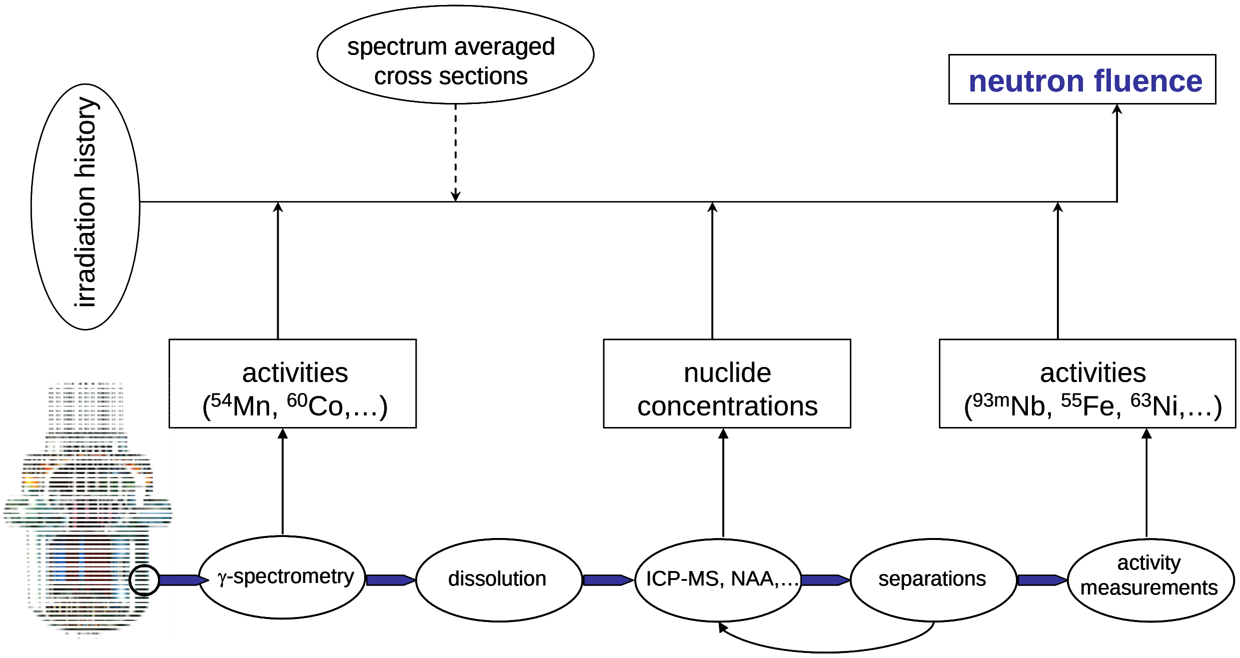
Objectives

Retrospective dosimetry needs a case by case evaluation. In the framework of a cooperation agreement between SCK•CEN (Belgium) and CNEA (Argentina), a feasibility study for performing retrospective dosimetry for Atucha I was performed.

The objective was to investigate experimental techniques for retrospective dosimetry using RPV steel samples irradiated in the BR2 reactor at SCK•CEN and samples coming from an Atucha I surveillance capsule unloaded in 1987.

Principal results

The general procedure for performing retrospective dosimetry is schematically shown in the figure below. A sample is obtained from structural material like the RPV. On this sample γ-spectrometry measurements are performed. Then the sample is chemically dissolved and the different nuclide concentrations are determined via for example ICP-MS or neutron activation analyses. The initial solution often contains overwhelming or interfering activities perturbing the γ-spectrometry (or β–) measurements and therefore several chemical separation procedures are applied to obtain sub-samples better suited for γ-spectrometry (or β–) measurements. The separation yields evidently have to be accurately determined, for example via ICP-MS. The combined information, nuclide concentrations and induced activities, together with the sample irradiation history, allow determining the incurred neutron fluence. As neutron induced cross sections are needed to convert specific activities to neutron fluence, the neutron spectrum has to be known or assumptions need to be made.



The measurements and results performed on and obtained from both types of Atucha I samples are chronologically summarised below. As the irradiation history of the samples from the surveillance capsule is not known, the neutron fluences are only determined for the samples irradiated in BR2. The neutron fluences accumulated by these samples are known from neutron dosimetry measurements which allows validating the retrospective method.

In a first step γ-spectrometry measurements were performed to determine the ^{60}Co and ^{54}Mn activity of both sample types. Due to the relatively short half-life of ^{54}Mn, this activity was only measurable on the samples irradiated in BR2. Then the samples were chemically dissolved and their isotopic composition was determined with ICP-MS.

No measurable Nb quantities were detected. From the initial solution separated Fe and Ni fractions were prepared and the separation yields were determined via ICP-MS (more than 80 % for Fe and 50 to 60 % for Ni).

The separated fractions were used for radioactivity measurements. From the Fe fractions thin deposits were prepared for measurement of the ^{55}Fe X-rays with a low-energy germanium detector relative to ^{55}Fe calibration samples. From the Ni fractions the ^{63}Ni β-activity was determined with liquid scintillation counting.

Finally the neutron fluences were calculated for the samples irradiated in BR2. The fast neutron fluence determination was based on the ^{54}Fe(n,p)^{54}Mn reaction. The results agree within 5 % with the expected value. The thermal neutron fluence determination was based on three different reactions: ^{59}Co(n,γ)^{60}Co, ^{54}Fe(n,γ)^{55}Fe and ^{62}Ni(n,γ)^{63}Ni. All results agree with the expected value within the estimated uncertainties. It can therefore be concluded that via retrospective dosimetry the fast and thermal neutron fluence can be determined from Atucha I steel if neutron spectrum and irradiation history information is available.

Future developments

The feasibility of applying retrospective dosimetry techniques for Atucha I has been demonstrated. If requested it will be possible to provide neutron dosimetry results in this way.
The technique of retrospective dosimetry can be further developed and standardised by investigating other types of material that are used as structural material in nuclear reactors.

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Main reference

J. Wagemans, J. Longhino, H. Blaumann and L. Adriaensen, in *Proceedings of the 13th International Symposium on Reactor Dosimetry*, 25-30 May 2008, Akersloot (The Netherlands), in preparation.